Claims

[c1] 1.A high pressure, high temperature capsule for containing at least one material and a solvent that becomes a supercritical fluid in a substantially air-free environment, said capsule comprising:

a)a closed end;

b)at least one wall adjoining said closed end and extending therefrom; and

c)a sealed end adjoining said at least one wall opposite said closed end,

wherein said at least one wall, said closed end, and said sealed end define a chamber therein for containing said at least one material and said solvent, wherein said capsule is formed from a deformable material, and wherein said capsule is fluid impermeable and chemically inert with respect to said at least one material and said supercritical fluid.

[c2] 2.The capsule according to Claim 1, wherein said capsule is formed from a cold-weldable material.

[c3] 3.The capsule according to Claim 2, wherein said cold-weldable material comprises at least one of copper, copper-based alloy, gold, silver, palladium, platinum, iridium, ruthenium, rhodium, osmium, iron, iron-based alloy, nickel, nickel-based alloy, and combinations thereof.

[c4] 4.The capsule according to Claim 1, wherein said deformable material comprises at least one of copper, copper-based alloy, gold, silver, palladium, platinum, iridium, ruthenium, rhodium, osmium, titanium, vanadium, chromium, iron, iron-based alloy, nickel, nickel-based alloy, zirconium, niobium, molybdenum, tantalum, tungsten, rhenium, and combinations thereof.

[c5] 5.The capsule according to Claim 1, further including at least one coating disposed on an inner surface of said capsule.

[c6] 6.The capsule according to Claim 5, wherein said at least one coating is formed from a first material comprising at least one of nickel, rhodium, gold, silver, palladium, platinum, ruthenium, iridium, tantalum, tungsten, rhenium, MC x N y O z , wherein M is at least one metal selected from aluminum, boron, silicon, titanium, vanadium, chromium, yttrium, zirconium, lanthanum, a rare earth

metal, hafnium, tantalum, tungsten, and wherein x, y, and z are between 0 and 3; and combinations thereof, and wherein said first material is different from said deformable material.

[c7] 7.The capsule according to Claim 5, wherein each of said at least one coating is between about 0.5 micron and about 250 microns in thickness.

[c8] 8.The capsule according to Claim 5, further including a diffusion barrier disposed between said inner surface and said at least one coating.

[c9] 9.The capsule according to Claim 8, wherein said diffusion barrier has a thickness of between about 10 nm and about 100 microns.

[c10] 10.The capsule according to Claim 8, wherein said diffusion barrier is formed from a second material comprising at least one of nickel, rhodium, platinum, palladium, iridium, ruthenium, rhenium, tungsten, molybdenum, niobium, silver, iridium, tantalum, MC x N y O z, where M is at least one metal selected from aluminum, boron, silicon, titanium, vanadium, chromium, yttrium, zirconium, lanthanum, a rare earth metal, hafnium, tantalum, tungsten, and x, y, and z are between 0 and 3; and combinations thereof, and wherein said second material is different from said first material and said deformable material.

[c11] 11.The capsule according to Claim 1, wherein said capsule further includes an outer capsule and an inner capsule nestingly disposed within said outer capsule and in a spaced apart relation to said outer capsule such that a free space exists between said outer capsule and said inner capsule, wherein each of said outer capsule and said inner capsule has at least one wall, a closed end, and a sealed end defining a chamber therein, and wherein said chamber of said inner capsule is adapted to contain said at least one material and said solvent.

[c12] 12.The capsule according to Claim 11, further including a pressure medium disposed in said free space, wherein said pressure medium equalizes a pressure within said inner capsule.

[c13] 13.The capsule according to Claim 11, further including a pressure medium

disposed in said free space, wherein the pressure medium provides an overpressure so that said at least one wall, said closed end, and said sealed end of said inner capsule are under one of compressive and neutral stress during processing at high pressure and high temperature.

[c14] 14.The capsule according to Claim 13, wherein said pressure medium comprises at least one of said solvent, water, ammonia, and carbon dioxide.

[c15] 15.The capsule according to Claim 11, wherein said inner capsule is formed from a glass.

[c16] 16.The capsule according to Claim 15, wherein said glass comprises at least one of fused quartz, fused silica, borosilicate glass, aluminosilicate glass, soda lime glass, soda barium glass, soda zinc glass, lead glass, potash soda lead glass, potash lead glass, or potash soda barium glass.

[c17] 17.The capsule according to Claim 11, wherein said inner capsule has a thickness of between about 0.1 mm and about 10 mm.

[c18] 18.The capsule according to Claim 1, further including an inert liner disposed on an inner surface of said at least one wall, said closed end, and said sealed end.

[c19] 19.The capsule according to Claim 18, wherein said inert liner has a thickness of between about 10 microns and about 5 mm.

[c20] 20.The capsule according to Claim 18, wherein said inert liner is formed from a first material comprising at least one of gold, platinum, rhodium, palladium, silver, iridium, ruthenium, osmium, tantalum, tungsten, rhenium, molybdenum, niobium, zirconium, yttrium, titanium, vanadium, chromium, silica, and combinations thereof, and wherein said first material is different from said deformable material.

[c21] 21.The capsule according to Claim 18, further including a diffusion barrier disposed between said inner surface and said inert liner.

[c22] 22.The capsule according to Claim 21, wherein said diffusion barrier has a

thickness of between about 10 nm and about 100 microns.

[c23] 23.The capsule according to Claim 21, wherein said diffusion barrier is formed from a second material comprising at least one of nickel, rhodium, platinum, palladium, iridium, ruthenium, rhenium, tungsten, molybdenum, niobium, silver, iridium, tantalum, MC x N y O z, wherein M is at least one metal selected from aluminum, boron, silicon, titanium, vanadium, chromium, yttrium, zirconium, lanthanum, a rare earth metal, hafnium, tantalum, tungsten, and x, y, and z are between 0 and 3; and combinations thereof, and wherein said second material is different from said first material and said deformable material.

[c24] 24.The capsule according to Claim 1, wherein each of said at least one wall, said closed end, and said sealed end has a thickness of between about 0.2 mm and about 10 mm.

[c25] 25.The capsule according to Claim 1, wherein said chamber is divided into two regions by a baffle.

[c26] 26.The capsule according to Claim 25, wherein said baffle has a fractional open area between about 0.5% and about 30%.

[c27] 27.The capsule according to Claim 25, wherein said baffle is formed from a first material and comprises at least one of copper, copper-based alloy, gold, silver, palladium, platinum, iridium, ruthenium, rhodium, osmium, titanium, vanadium, chromium, iron, iron-based alloy, nickel, nickel-based alloy, zirconium, niobium, molybdenum, tantalum, tungsten, rhenium, silica, alumina, and combinations thereof.

[c28] 28.The capsule according to Claim 25, further including at least one coating disposed on said baffle.

[c29] 29.The capsule according to Claim 28, wherein said at least one coating is formed from a second material comprising at least one of nickel, rhodium, gold, silver, palladium, platinum, ruthenium, iridium, tantalum, tungsten, rhenium, MC x N y O z, where M is at least one metal selected from aluminum, boron,

silicon, titanium, vanadium, chromium, yttrium, zirconium, lanthanum, a rare earth metal, hafnium, tantalum, tungsten, and x, y, and z are between 0 and 3; and combinations thereof, and wherein said second material is different from said first material.

[c30] 30.The capsule according to Claim 1, wherein said sealed end comprises a lid having a sealed fill tube, wherein said lid is sealed to said at least one wall by one of a pipe thread seal, a metal-to-metal compression seal, a gasket seal, and a weld seal.

[c31] 31.The capsule according to Claim 30, wherein said lid and said fill tube comprise at least one of copper, copper-based alloy, gold, silver, palladium, platinum, iridium, ruthenium, rhodium, osmium, titanium, vanadium, chromium, iron, iron-based alloy, nickel-based alloy, zirconium, niobium, molybdenum, tantalum, tungsten, rhenium, silica, alumina, or combinations thereof.

[c32] 32.The capsule according to Claim 1, further including an outer seal joined to said sealed end of said capsule.

[c33] 33.The capsule according to Claim 32, wherein said outer seal surrounds said capsule in its entirety.

[c34] 34.The capsule according to Claim 32, wherein said outer seal is formed from at least one of copper, copper alloy, gold, silver, palladium, platinum, iridium, ruthenium, rhodium, osmium, titanium, vanadium, chromium, nickel, nickel alloy, iron, steel, iron alloy, zirconium, niobium, molybdenum, tantalum, tungsten, rhenium, and combinations thereof.

[c35] 35.The capsule according to Claim 1, wherein said capsule is impermeable to at least one of hydrogen, oxygen, and nitrogen.

[c36] 36.The capsule according to Claim 1, wherein said capsule is self-pressurizing.

[c37] 37.The capsule according to Claim 36, wherein said capsule is self-pressurizing from about 1 bar up to about 80 kbar.

[c38] 38.The capsule according to Claim 37, wherein said capsule is self-pressurizing up to between about 5 kbar and about 80 kbar.

[c39] 39.The capsule according to Claim 38, wherein said capsule is self-pressurizing up to between about 5 kbar and about 60 kbar.

[c40] 40.A plug for sealing a high pressure, high temperature capsule for containing at least one material and a solvent that becomes a supercritical fluid in a substantially air-free environment, said capsule having at least one wall, a closed end, and a sealed end defining a chamber therein for containing said at least one material and said solvent, said plug comprising a cold-weldable material and being sealingly insertable in an open end of said capsule, wherein said sealed end is formed by inserting said plug in said open end and cold welding said plug to said capsule.

[c41] 41.The plug according to Claim 40, wherein said cold-weldable material comprises at least one of copper, copper-based alloy, gold, silver, palladium, platinum, iridium, ruthenium, rhodium, osmium, iron, iron-based alloy, nickel, nickel-based alloy, and combinations thereof.

[c42] 42.The plug according to Claim 40, further including at least one coating disposed on an inner surface of said plug.

[c43] 43.The plug according to Claim 40, wherein said at least one coating is formed from a first material and comprises at least one of nickel, rhodium, gold, silver, palladium, platinum, ruthenium, iridium, tantalum, tungsten, rhenium, $MC_xN_yO_z$, wherein M is at least one metal selected from aluminum, boron, silicon, titanium, vanadium, chromium, yttrium, zirconium, lanthanum, a rare earth metal, hafnium, tantalum, tungsten, and x, y, and z are between 0 and 3; and combinations thereof, and wherein said first material is different from said cold-weldable material.

[c44] 44.The plug according to Claim 40, further including a fill tube joined to said plug, wherein said fill tube has an orifice that extends through said plug to an inner surface of said plug.

[c45] 45.A high pressure, high temperature capsule for containing at least one material and solvent that becomes a supercritical fluid in a substantially air-free environment, said capsule comprising:

a)a closed end;

b)at least one wall adjoining said closed end and extending therefrom; and

c)a sealed end adjoining said at least one wall opposite said closed end, said sealed end comprising a plug that is cold-welded to said at least one wall, wherein said at least one wall, said closed end, and said sealed end define a chamber therein for containing said at least one material and said solvent, wherein said capsule is formed from a deformable cold-weldable material, and wherein said capsule is fluid impermeable and chemically inert with respect to said at least one material and said supercritical fluid.

[c46] 46.The capsule according to Claim 45, wherein said deformable cold-weldable material comprises at least one of copper, copper-based alloy, gold, silver, palladium, platinum, iridium, ruthenium, rhodium, osmium, iron, iron-based alloy, nickel, nickel-based alloy, and combinations thereof.

[c47] 47.The capsule according to Claim 45, further including at least one coating disposed on an inner surface of said capsule.

[c48] 48.The capsule according to Claim 47, wherein said at least one coating is formed from a first material comprising at least one of nickel, rhodium, gold, silver, palladium, platinum, ruthenium, iridium, tantalum, tungsten, rhenium, MC xN yO z, wherein M is at least one metal selected from aluminum, boron, silicon, titanium, vanadium, chromium, yttrium, zirconium, lanthanum, a rare earth metal, hafnium, tantalum, tungsten, and x, y, and z are between 0 and 3; and combinations thereof, and wherein said first material is different from said deformable cold-weldable material.

[c49] 49.The capsule according to Claim 47, wherein each of said at least one coating is between about 0.5 micron and about 250 microns in thickness.

[c50] 50.The capsule according to Claim 47, further including a diffusion barrier disposed between said inner surface and said at least one coating.

[c51] 51.The capsule according to Claim 50, wherein said diffusion barrier has a thickness of between about 10 nm and about 100 microns.

[c52] 52.The capsule according to Claim 50, wherein said diffusion barrier is formed from a second material comprising at least one of nickel, rhodium, platinum, palladium, iridium, ruthenium, rhenium, tungsten, molybdenum, niobium, silver, iridium, tantalum, MC x N y O z, where M is at least one metal selected from aluminum, boron, silicon, titanium, vanadium, chromium, yttrium, zirconium, lanthanum, a rare earth metal, hafnium, tantalum, tungsten, and x, y, and z are between 0 and 3; and combinations thereof, and wherein said second material is different from said first material and said deformable cold-weldable material.

[c53] 53.The capsule according to Claim 45, wherein said capsule further includes: an outer capsule and an inner capsule nestingly disposed within said outer capsule and in a spaced apart relation to said outer capsule such that a free space exists between said outer capsule and said inner capsule, wherein each of said outer capsule and said inner capsule has at least one wall, a closed end, and a sealed end defining a chamber therein, and wherein said chamber of said inner capsule is adapted to contain said at least one material and said solvent.

[c54] 54.The capsule according to Claim 53, further including a pressure medium disposed in said free space, wherein said pressure medium equalizes a pressure within said inner capsule.

[c55] 55.The capsule according to Claim 53, further including a pressure medium disposed in said free space, wherein the pressure medium provides an overpressure so that said at least one wall, said closed end, and said sealed end of said inner capsule are under one of compressive stress and neutral stress during processing at high pressure and high temperature.

[c56] 56.The capsule according to Claim 55, wherein said pressure medium comprises at least one of the solvent contained within the inner capsule, water, ammonia, or carbon dioxide.

[c57] 57.The capsule according to Claim 53, wherein said inner capsule is formed

from a glass.

[c58] 58.The capsule according to Claim 57, wherein said glass comprises at least one of fused quartz, fused silica, borosilicate glass, aluminosilicate glass, soda lime glass, soda barium glass, soda zinc glass, lead glass, potash soda lead glass, potash lead glass, and potash soda barium glass.

[c59] 59.The capsule according to Claim 53, wherein said inner capsule has a thickness of between about 0.1 mm and about 10 mm.

[c60] 60.The capsule according to Claim 45, further including an inert liner disposed on an inner surface of said at least one wall, said closed end, and said sealed end.

[c61] 61.The capsule according to Claim 60, wherein said inert liner has a thickness of between about 10 microns and about 5 mm.

[c62] 62.The capsule according to Claim 60, wherein said inert liner is formed from a first material comprising at least one of gold, platinum, rhodium, palladium, silver, iridium, ruthenium, osmium, tantalum, tungsten, rhenium, molybdenum, niobium, zirconium, yttrium, titanium, vanadium, chromium, silica, and combinations thereof, wherein said first material is different from said deformable cold-weldable material.

[c63] 63.The capsule according to Claim 60, further including a diffusion barrier disposed between said inner surface and said inert liner.

[c64] 64.The capsule according to Claim 63, wherein said diffusion barrier has a thickness of between about 10 nm and about 100 microns.

[c65] 65.The capsule according to Claim 63, wherein said diffusion barrier is formed form a second material comprising at least one of nickel, rhodium, platinum, palladium, iridium, ruthenium, rhenium, tungsten, molybdenum, niobium, silver, iridium, tantalum, MC x N y O z, where M is at least one metal selected from aluminum, boron, silicon, titanium, vanadium, chromium, yttrium, zirconium, lanthanum, a rare earth metal, hafnium, tantalum, tungsten, and x, y, and z are between 0 and 3; and combinations thereof, and wherein said first

material is different from said second material and said deformable cold-weldable material.

[c66] 66.The capsule according to Claim 45, wherein said at least one wall, said closed end, and said sealed end each have a thickness of between about 0.2 mm and about 10 mm.

[c67] 67.The capsule according to Claim 45, wherein said chamber is divided into two regions by a baffle.

[c68] 68.The capsule according to Claim 67, wherein said baffle has a fractional open area between about 0.5% and about 30%.

[c69] 69.The capsule according to Claim 67, wherein said baffle is formed from a first material comprising at least one of copper, copper-based alloy, gold, silver, palladium, platinum, iridium, ruthenium, rhodium, osmium, titanium, vanadium, chromium, iron, iron-based alloy, nickel, nickel-based alloy, zirconium, niobium, molybdenum, tantalum, tungsten, rhenium, silica, alumina, and combinations thereof.

[c70] 70.The capsule according to Claim 67, further including at least one coating disposed on said baffle.

[c71] 71.The capsule according to Claim 67, wherein said at least one coating is formed from a second material comprising at least one of nickel, rhodium, gold, silver, palladium, platinum, ruthenium, iridium, tantalum, tungsten, rhenium, $MC_xN_yO_z$, where M is at least one metal selected from aluminum, boron, silicon, titanium, vanadium, chromium, yttrium, zirconium, lanthanum, a rare earth metal, hafnium, tantalum, tungsten, and x, y, and z are between 0 and 3; and combinations thereof, and wherein said second material is different from said first material.

[c72] 72.The capsule according to Claim 45, wherein said sealed end comprises a lid having a sealed fill tube, wherein said lid is sealed to said at least one wall by one of a pipe thread seal, a metal-to-metal compression seal, a gasket seal, and a weld seal.

[c73] 73.The capsule according to Claim 72, wherein said lid and said fill tube comprise at least one of copper, copper-based alloy, gold, silver, palladium, platinum, iridium, ruthenium, rhodium, osmium, titanium, vanadium, chromium, iron, iron-based alloy, nickel-based alloy, zirconium, niobium, molybdenum, tantalum, tungsten, rhenium, silica, alumina, or combinations thereof.

[c74] 74.The capsule according to Claim 45, further including an outer seal joined to said sealed end of said capsule.

[c75] 75. The capsule according to Claim 74, wherein said outer seal surrounds said capsule in its entirety.

[c76] 76.The capsule according to Claim 74, wherein said outer seal is formed from at least one of copper, copper alloy, gold, silver, palladium, platinum, iridium, ruthenium, rhodium, osmium, titanium, vanadium, chromium, nickel, nickel alloy, steel, iron, iron alloy, zirconium, niobium, molybdenum, tantalum, tungsten, rhenium, and combinations thereof.

[c77] 77.The capsule according to Claim 45, wherein said capsule is self-pressurizing.

[c78] 78.The capsule according to Claim 77, wherein said capsule is self-pressurizing from about 1 bar up to about 80 kbar.

[c79] 79.The capsule according to Claim 78, wherein said capsule is self-pressurizing up to between about 5 kbar and about 80 kbar.

[c80] 80.The capsule according to Claim 79, wherein said capsule is self-pressurizing up to between about 5 kbar and about 60 kbar.

[c81] 81.A method of filling a high pressure, high temperature capsule with at least one material and a solvent that becomes a supercritical fluid in a substantially air-free environment, said capsule having at least one wall, a closed end, and an open sealable end defining a chamber therein for containing said at least one material and said supercritical fluid, the method comprising the steps of:

a)providing the capsule;

b)providing the at least one material to the chamber;

c)providing a solvent source, wherein the solvent source contains the solvent and is connectable to a vacuum manifold;

d)connecting the solvent source to the vacuum manifold;

e)inserting the at least one material into the chamber;

f)placing the chamber of the capsule in fluid communication with the vacuum manifold and evacuating the chamber to a predetermined pressure;

g)cooling the chamber to a temperature below a predetermined temperature;

h)placing the chamber and the solvent source in communication with each other through the vacuum manifold; and;

i)providing a portion of the solvent into the chamber, thereby filling the open-ended capsule to a predetermined level.

[c82] 82.The method of Claim 81, wherein the step of placing the chamber of the capsule in fluid communication with the vacuum manifold and evacuating the chamber to a predetermined pressure comprises placing the chamber of the capsule in fluid communication with the vacuum manifold and evacuating the chamber to a pressure of less than about 1 torr.

[c83] 83.The method of Claim 81, wherein the step of cooling the chamber to a temperature below a predetermined temperature comprises cooling the chamber to a temperature at which the solvent has a vapor pressure of less than about 760 Torr.

[c84] 84.The method of Claim 81, wherein the step of providing a portion of the solvent into the chamber comprises condensing a portion of the solvent into the chamber.

[c85] 85.The method of Claim 84, wherein the step of condensing a portion of the solvent into the chamber comprises controlling a mass flow of the solvent into the chamber for a predetermined time period and condensing the solvent in the chamber.

[c86] 86. The method of Claim 81, wherein the step of providing a portion of the solvent into the chamber comprises:

a)providing a portion of the solvent at an initial predetermined pressure to a predetermined volume;
b)condensing a portion of the solvent into the chamber; and measuring a final pressure in the predetermined volume.

[c87] 87.The method of Claim 81, wherein the step of providing a portion of the solvent into the chamber comprises injecting a portion of the solvent into the chamber.

[c88] 88.A method of sealing a high pressure, high temperature capsule containing at least one material and a solvent that becomes a supercritical fluid at high temperature and high pressure in a substantially air-free environment, said capsule having at least one wall, a closed end, and an open sealable end defining a chamber therein for containing said at least one material and said solvent, the method comprising the steps of:
a)providing the capsule containing the at least one material;
b)placing the chamber of the capsule in communication with a vacuum manifold and evacuating the chamber to a predetermined pressure;
c)filling the chamber with a predetermined quantity of the solvent; and
d)sealing the open sealable end of the capsule.

[c89] 89.The method of Claim 88, wherein the step of sealing the open sealable end of the capsule comprises:
a)applying heat to a portion of the at least one wall at the open sealable end of the capsule;
b)collapsing the portion of the at least one wall at the open sealable end; and
c)forming a solid weld, thereby sealing the open sealable end of the capsule.

[c90] 90.The method of Claim 88, wherein the step of sealing the open sealable end of the capsule comprises one of torch welding, arc welding, ultrasound welding, and vibratory welding a portion of the at least one wall at the open sealable end of the capsule.

[c91] 91.The method of Claim 88, wherein the step of sealing the open sealable end of the capsule comprises:

a)applying pressure to an outer surface of a portion of the at least one wall at the open sealable end of the capsule;

b)collapsing the portion of the at least one wall at the open sealable end; and

c)forming a cold weld, thereby sealing the open sealable end of the capsule.

[c92] 92.The method of Claim 88, wherein the step of sealing the open sealable end of the capsule comprises:

a)inserting a plug into the open sealable end of the capsule;

b)contacting the at least one wall with the plug;

c)applying pressure to an outer surface of at least one of the at least one wall and the plug; and

d)forming a cold weld between the at least one wall and the plug, thereby sealing the open sealable end of the capsule.

[c93] 93.An apparatus for sealing a high pressure, high temperature capsule with a cold-weldable plug to a form a substantially air-free chamber within said capsule, said apparatus comprising:

a)a movable ram for inserting said cold-weldable plug into an open sealable end of said capsule;

b)a mechanical support for supporting said capsule and guiding said ram, wherein said mechanical support and said ram form an air-tight inner chamber; and

c)a vacuum inlet passing through said mechanical support to said air-tight inner chamber, wherein said vacuum inlet provides communication between said air-tight inner chamber and a vacuum manifold,

wherein said cold-weldable plug is cold-welded to at least one wall of said capsule when said cold-weldable plug is inserted into said open sealable end and a pressure is applied to said ram.

[c94] 94.A gallium nitride single crystal, wherein the gallium nitride single crystal is formed by: providing at least one gallium nitride source material to a high pressure, high temperature capsule, the capsule having at least one wall, a closed end, and an open sealable end defining a chamber therein for containing said at least one gallium nitride source material and a solvent that becomes a

supercritical fluid at high temperature and high pressure; placing the chamber of the capsule in fluid communication with a vacuum manifold and evacuating the chamber to a predetermined pressure; filling the chamber with a predetermined quantity of the solvent; sealing the open sealable end of the capsule; disposing the sealed capsule within a pressure vessel comprising a pressure transmission medium surrounding the capsule for maintaining an outer pressure on the capsule, a heating element insertable in the pressure transmission medium such that the heating element surrounds the capsule, a restraint to contain and hold in place the capsule, the pressure transmission medium, the heating element, and at least one seal between the restraint and the pressure transmission medium; and subjecting the capsule to high pressure, high temperature conditions, wherein the solvent contained within the sealed capsule becomes a supercritical fluid and generates a predetermined pressure within the sealed capsule, and wherein the supercritical fluid reacts with the at least one gallium nitride source material to form the gallium nitride single crystal.